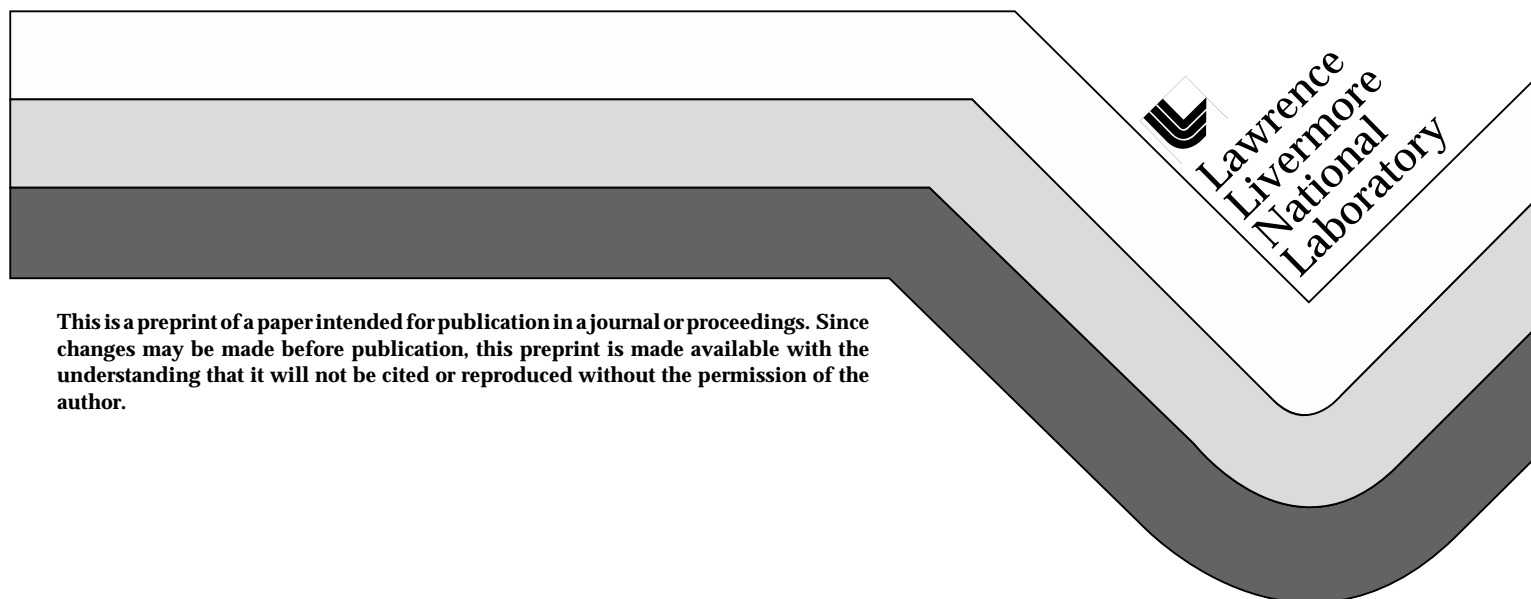


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C. R. Wuest
B. A. Fuchs
X. Shi

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PRECISION MACHINING, POLISHING AND MEASUREMENT OF MECHANICAL AND TOXICOLOGICAL PROPERTIES OF LEAD TUNGSTATE CRYSTALS FOR THE CMS ELECTROMAGNETIC CALORIMETER

Craig R. Wuest, Baruch A. Fuchs, Xiaorong Shi
Lawrence Livermore National Laboratory
P.O. Box 808
Livermore, CA 94551 USA

We have developed new machining and polishing techniques that have previously been applied to large scintillating crystal arrays for high energy physics experiments such as the Barium Fluoride Electromagnetic Calorimeter for the GEM Detector at SSCL, the CsI Electromagnetic Calorimeter for the BaBar Detector at PEP-II B Factory at SLAC and the 110,000 crystal CMS Lead Tungstate Electromagnetic Calorimeter at LHC at CERN. We discuss earlier results achieved with diamond machining and polishing methods and present new results on diamond machining of lead tungstate crystals. Additionally we present new results on mechanical properties of lead tungstate including toxicological data important for the safe handling and processing of this material.

INTRODUCTION

New machining and polishing techniques have been developed at Lawrence Livermore National Laboratory (LLNL) for large scintillating crystals that optimize crystalline surfaces while minimizing sub-surface damage or stress. Surface optimization is critical to ensure long term stability of crystals in calorimeter applications where they may be subject to many years of radiation exposure, temperature cycling and confinement without the ability to replace or repair degraded or damaged crystals. Our studies on barium fluoride crystals produced fully crystal surfaces using diamond-loaded pitch lapping techniques [1]. These crystalline surfaces were verified by Atomic Force Microscopy (AFM) and Rutherford Back-scattering (RBS) analyses. Surface roughness of about 10-20 angstroms and sub-micron mechanical tolerances were demonstrated on large crystals. Mass reduction techniques were also developed for machining and polishing up to five 50 cm long crystals at one time. The technology was found to be applicable for a number of new crystal detectors proposed at high energy physics colliders including most recently the Compact Muon Solenoid Detector Lead Tungstate Electromagnetic Calorimeter (CMS ECAL) at LHC [2].

LLNL has an active program of study on lead tungstate (PWO) as part of our responsibility within the CMS ECAL Program. This includes the development of a specialized cutting and polishing machine for proper characterization of PWO boules immediately after removal from the furnaces and prior to cutting the crystals to their final long, tapered trapezoidal shapes. Crystals are to be inspected using a transmissometer to ensure the requisite optical quality. In addition the end-faces of the crystal need to be polished to a high quality to enhance light collection properties, which are dependent on high reflectivity mirrored front and rear surfaces. These mirrored surfaces might be applied in the form of a reflective coating, in which case the quality of the surface is critical for the long term stability of the coating. Additionally a polished optically flat surface is required for a high quality bonding of the photodetector to the crystal (the CMS ECAL will use avalanche photodiodes). We present work on machining PWO crystals applicable to the development of this special endface cutting and polishing machine.

PWO machining produces PWO wastes, typically fine chips of the material mixed into cutting oil and solvents. We are concerned with the safety of the personnel who will come into contact with these waste products as well as the environmental implications of disposal of lead-containing wastes. In order to provide quantitative answers to the question of lead contamination of personnel, machines and the environment we have begun a program of study of PWO solubility in cutting oils.

LLNL is working in collaboration with ECA collaboration members at CERN and elsewhere to develop the thermal model of the ECAL along with the proper monitoring system and we are also working to design the support structure for the large number of crystals. Because of this responsibility we are making measurements of fundamental crystal properties in order to properly understand the thermal and mechanical behavior of the CMS ECAL.

LEAD TUNGSTATE SURFACE PREPARATION AND ANALYSIS

Surface preparation is critical to the performance of crystals for a number of reasons. First, an improperly prepared (machined, ground, polished, lapped) crystal suffers from induced stresses and deformations in the first few hundred microns of the surface. These stresses can manifest themselves in the formation of cracks (crazing) over long times, or more quickly when subjected to extremes of heat, radiation, humidity, etc.. Surface stresses can be minimized using well-known polishing and lapping techniques that *gently* bring the surface to a final finish. These techniques have been developed at LLNL for barium fluoride and also applied to cerium fluoride, lead fluoride and

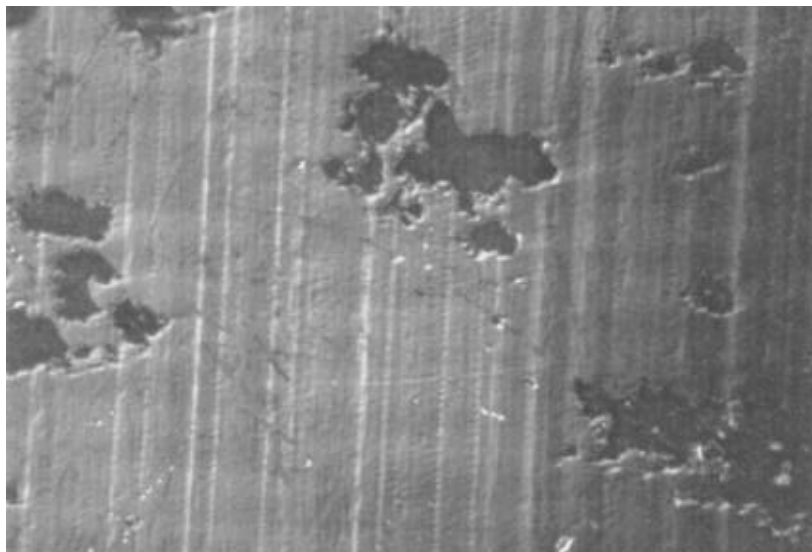


Figure 1. Photograph (179x) of single point diamond turning of PWO showing regions of both plastic flow and fracture.

PWO. Improper surface preparation can introduce contaminants into the surface of the crystal. Under certain conditions these contaminants can migrate into the bulk of the crystal and cause areas of radiation susceptibility. Because these scintillator materials emit their light typically in the blue or near UV, surface finish is especially important for good light transport properties.

We have previously described surface preparation techniques explored at LLNL including ion beam milling, diamond turning with a single point diamond tool, and various polishing/lapping techniques [1]. In general we have found that ion beam milling provides the best crystalline surface, however, the uniformity of the surface, as well as the surface finish is not very good. In terms of surface finish, diamond-turned surface finishes can be quite impressive –6 Å RMS finishes have been demonstrated on barium fluoride. However, RBS analysis of diamond-turned surfaces reveal that they are amorphous, presumably due to plastic flow of the material under the action of the diamond tool. Figure 1 shows a diamond turned sample of PWO. We observe that the material exhibits localized areas of plastic flow interspersed with areas of fracture. We conjecture that improved PWO growing and annealing methods might lead to fully diamond-turnable crystal surfaces.

In the meantime another polishing technique – pitch lapping with diamond abrasives – provides the best combination of surface polish (10-20 Å RMS) and surface crystallinity for most crystals. The technique is applied after more standard polishing techniques and is a simple lap prepared with a low melting temperature synthetic pitch. Grooves are formed in the pitch in a pattern to allow cutting fluids, abrasives and ground material to be washed away during the lapping process. The key to the process is a final polish with an abrasive of very uniformly sized diamond, 0.5 μm or 0.25 μm diameter, imbedded in the pitch. In addition, a non-aqueous cutting fluid such as low viscosity oil, or ethylene glycol is used to uniformly disperse the diamond and to carry away waste material. Water is not a good fluid for diamond polishing because of the tendency of diamond to agglomerate in water. These polishing techniques are simple to implement and are essentially extensions of standard polishing techniques already in practice in the US and elsewhere. These techniques are easily transferred to industry both in the US and abroad. LLNL engineers and physicists have successfully worked with our counterparts at the Shanghai Institute of Ceramics and the Beijing Glass Research Institute to develop this capability [3]. Figure 2 shows examples of improper (as received from the crystal grower) and proper diamond-loaded pitch lap) polishing of PWO.

MEASUREMENTS OF LEAD TUNGSTATE MECHANICAL PROPERTIES

Measurements of PWO mechanical properties are being made at LLNL to better define the CMS ECAL structural and thermal systems. We have measured the coefficient of thermal expansion of a small ~1 cm x 1 cm x 0.5 cm crystal sample cut from a larger crystal. Measurements were made using a DuPont 9900 Thermal Mechanical Analysis system and the sample was cycled from dry ice temperature (-38 degrees Centigrade) to +100 degrees Centigrade. In the sample we prepared, the crystal's principle, or "a" axis is about 19 degrees off from the actual growth axis, which corresponds to the direction of the 0.5 cm sample dimension. Thus we measure a superposition of components of linear expansion along the two principle axes of the crystal. The coefficient of thermal expansion is linear in the range of 15 degrees to 30 degrees, the range of expected minimum and maximum temperature in the ECAL. The results give a coefficient of thermal expansion of $10.0 \times 10^{-6}/^{\circ}\text{C}$ and $11.5 \times 10^{-6}/^{\circ}\text{C}$ for the two 1 cm long sides in the range of 15-30 $^{\circ}\text{C}$. This can be compared with measurements from the Shanghai Institute of Ceramics which give $9.58 \times 10^{-6}/^{\circ}\text{C}$ at 100 $^{\circ}\text{C}$ perpendicular to

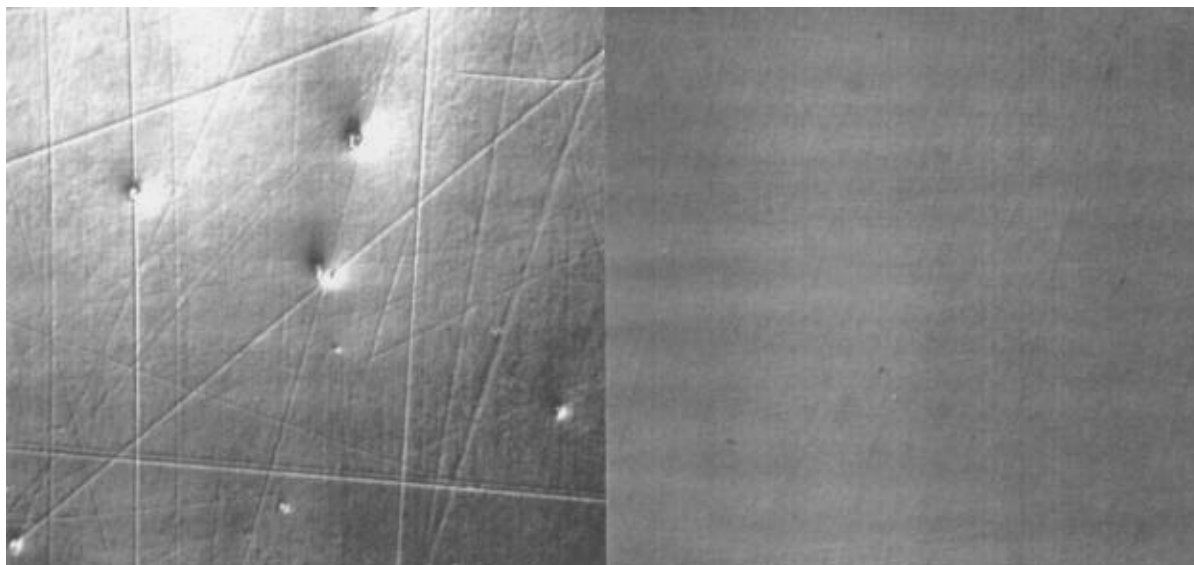


Figure 2. Photograph (x179) of two polished PWO surfaces. On the left is the as-received crystal with large surface scratches and inclusions of polishing debris. poor surfaces such as this exhibit large amounts of subsurface damage and stress. The large scratches and pits make application of high quality reflective coatings difficult. on the right is a PWO surface polished at LLNL using diamond-loaded pitch lapping. The surface roughness is $\sim 15\text{-}20 \text{ \AA RMS}$.

the “a” axis and $22.7 \times 10^{-6}/^{\circ}\text{C}$ parallel to the “a” axis. Earlier work by Krishna Rao and Deshpande give $8.13 \times 10^{-6}/^{\circ}\text{C}$ and $19.73 \times 10^{-6}/^{\circ}\text{C}$ perpendicular and parallel to the “a” axis, respectively, at 30°C [4]. Our result, measured perpendicular to the growth axis and 19 degrees offset from the “a” axis is consistent with previous measurements.

We have measured the Young's Modulus of compression of a right circular cylinder of PWO with dimensions 1 cm in diameter and 2 cm long. The sample was prepared with the cylinder axis in the direction of the growth axis of the crystal from which it was taken. To prevent stress from building up in localized regions of the crystal due to non-uniform loading, the sample was prepared as a cylinder with very parallel faces. The sample was measured on an Instron Model 1125, 20k pound test machine using a 20k pound load cell set to the 500 pound load range. A spherical seat was installed at the bottom platen to correct for any remaining deviation from parallelism between the top and bottom surfaces. Three MTS 0.3 inch 15% extensometers were installed on the test sample to provide average strain data. Figure 3 shows the setup. Three runs were made at room temperature with a constant crosshead travel of 0.002 inches (50 microns)/minute. The first run loaded the sample to 250 pounds. The second run inadvertently began with the crosshead loading the sample to 2100 pounds prior to the error being detected. The sample developed a slight internal fracture but did not break. The third run took the sample to 500 pounds to determine if the modulus had changed. The results of the third run are shown in Figure 3. No large change in modulus was observed between the first and third runs and the results of the modulus were measured to be 9.0×10^6 psi and 9.27×10^6 psi (62.1 GPa and 64.0 GPa), respectively. For comparison the modulus of aluminum alloy is 10×10^6 psi and lead silicate glass is 9×10^6 psi. Our result is comparable to measurements made by researchers at CERN and Ecole Polytechnique [5].

MEASUREMENTS OF LEAD SOLUBILITY FROM LEAD TUNGSTATE

We have also made measurements of lead solubility in cutting oil. Two experiments have been performed. The first (qualitative) measurement was made using chips of PWO mixed with hydrocarbon cutting oil (Drakeol 7) used in the diamond turning study. The sample included 0.5 cc of chips ranging in size from about 1-20 microns. The chips were immersed in 20 cc of cutting oil. The sample was aged for 43 days at room temperature and then analyzed. In the analysis the cutting oil was separated from the solid material by centrifugation at 4000 rpm for 10 minutes. 0.2 gram of oil was then wet ashed in concentrated $\text{HNO}_3/\text{H}_2\text{SO}_4$ in a closed Teflon digestion vessel to destroy any organic material. The total volume of material was then evaporated to approximately 0.5 g to which 100 ng of thallium was added as an internal standard. Following volume adjustment to a final volume of 10 ml, the presence of lead was determined by inductively coupled plasma mass spectrometry (ICP-MS). A clean sample of unused cutting oil was also measured in the same manner for comparison. The results show at the amount of lead is 160 ng/g for the unexposed oil and 530 ng/g for the exposed oil. Both measurements have an uncertainty of about 10%.

In a second ongoing quantitative experiment a PWO cube, 1 cm on a side, is being continuously exposed to cutting oil in a polyethylene bottle. Small amounts of exposed and unexposed oil will be measured over regular periods approximately once per month for the next few months. Occupational

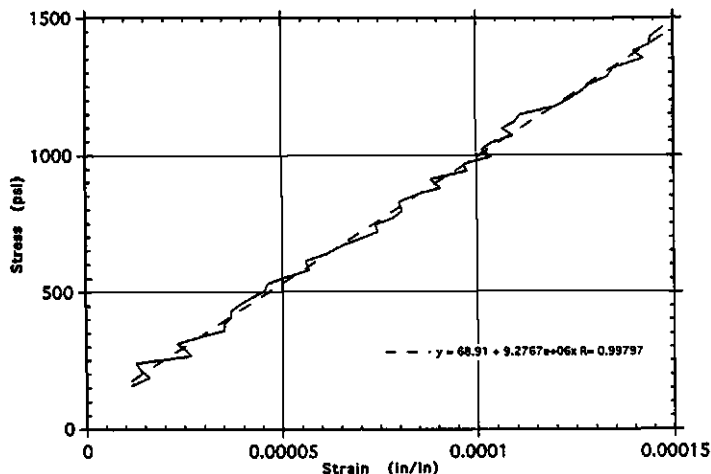


Figure 3. Photograph of the Young's Modulus test setup and results of a 500 pound load run.

health and safety regulations at LLNL do not specify a limit on skin contact with lead because elemental lead is not readily absorbed through the skin. Exposure limits are available for ingestion of lead. The US EPA limit for lead exposure in drinking water is 15 ng/g. Environmental regulations at LLNL also define lead wastes as containing fewer than 200 ng/g of lead [6]. The oil waste can also contain organo-lead compounds, which might be absorbed through the skin or inhaled. Thus we see that the PWO solubility in cutting oil poses a problem for handling and disposal by LLNL standards. Proper collection, processing and disposal of this lead-containing hydrocarbon oil waste is necessary. Protective gloves should be used to prevent ingestion via hand to mouth.

CONCLUSIONS

LLNL is contributing to the understanding of PWO as part of the effort to build the CMS ECAL sub-system. We have seen that PWO is a material that challenges our abilities to produce a high quality electromagnetic calorimeter. Our diverse program of study encompasses many aspects important to this effort including materials properties, machining, polishing, fundamental physical constants and personnel and environmental safety. We intend to continue this program of study during the R&D phase of CMS to develop machines for production factories and to provide input to mechanical and thermal models of the ECAL.

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Technical Information Department • Lawrence Livermore National Laboratory
University of California • Livermore, California 94551

